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AUTHOR(S): F. A. Guevara
R. F. Dvorak

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AN OVERVIEW OF AIRBORNE RADIOACTIVE EMISSIONS AT LOS ALAMOS NATIONAL LABORATORY

F. A. Guevara, M.S. R. F. Dvorak, M.S.

ABSTRACT

Strict control is essential over any emissions of radioactivity in the ventilation exhaust from facilities where radioactive materials may become airborne. At Los Alamos National Laboratory there are 87 stacks exhausting ventilation air to the environment from operations with a potential for radioactive emissions. These stacks cover the diverse operations at all Laboratory facilities where radioactive materials are handled and require continuous sampling/monitoring to detect levels of contamination. An overview is presented of the operations, associated ventilation exhaust clean up systems, and analysis of the emissions. In keeping with the as-low-as-reasonably-achievable concept, emissions of radionuclides are reduced whenever practicable. A specific example describing the reduction of emissions from the linear accelerator beam stop area at the Los Alamos Meson Physics Facility during 1985 by a factor of 8 over previous emissions is presented.

Key Words

Emissions, radioactivity, ventilation, exhaust, environment, sampling, monitoring, reduction

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Strict control is essential over any emissions of radioactivity in the ventilation exhaust from facilities where radioactive materials may become airborne. At Los Alamos National Laboratory there are 87 stacks exhausting ventilation air to the environment from operations with a potential for radioactive emissions. These stacks cover the diverse operations at all Laboratory facilities where radioactive materials are handled and require continuous sampling/monitoring to detect levels of contamination. An overview is presented of the operations, associated ventilation exhaust clean up systems, and analysis of the emissions. In keeping with the as-low-as-reasonably-achievable concept, emissions of radionuclides are reduced whenever practicable. A specific example describing the reduction of emissions from the linear accelerator beam stop area at the Los Alamos Meson Physics Facility during 1985 by a factor of 8 over previous emissions is presented.

INTRODUCTION

Currently 87 ventilation exhaust points at the Los Alamos National Laboratory have a potential for releasing radioactively contaminated air to the atmosphere. The emission of trace quantities of airborne nuclides from the diverse research and development operations at Los Alamos is closely controlled and monitored in accordance with Department of Energy (DOE) orders (DOE 1981) and Environmental Protection Agency (EPA) standards (EPA 1985).

F. A. Guevara, Staff Member, Radiation Protection Group, and R. F. Dvorak, Staff Member, Accelerator Health Protection Group, Los Alamos National Laboratory.

The air cleaning systems installed at Los Alamos to control contamination vary from the familiar banks of High Efficiency Particulate Air (HEPA) filters to custom designed tritium removal systems and include most of the emission control systems commonly used throughout industry. In this paper a summary report is given of the potential sources of airborne radioactivity at Los Alamos and their control, sampling, measurement, and documentation. An example of reducing emissions in adherence to the concept of as-low-as-reasonably (ALARA) is presented. The example describes a modification made at the beam stop area of the Los Alamos Meson Physics Accelerator that resulted in a significant lowering of radioactive emissions from this operation.

EMISSIONS AT THE LOS ALAMOS NATIONAL LABORATORY

The Laboratory is situated in Los Alamos County, north central New Mexico, approximately 60 mi (100 km) NE of Albuquerque (Fig. 1) and is operated for the DOE by contract with the University of California. Since its begining in 1943 the primary mission has been nuclear weapons research and development. The spectrum of programs includes magnetic and inertial fusion, nuclear fission, nuclear safeguards and security, and laser isotope separation. Historically basic research in the areas of physics, chemistry, and engineering to support national defense programs has always been a major part of the Laboratory's efforts. In addition the Laboratory is assigned research on peaceful uses of nuclear energy including space applications, power reactor projects, radiobiology, medicine, and radioactive waste management. Most of these programs involve the handling of nuclear materials and thus there is a significant potential for the emission of airborne radionuclides to the environment.

(Figure 1. Location of Los Alamos National Laboratory.)

During operations in 1986 emissions of radionuclides were routinely monitored from 87 ventilation exhaust points at the Laboratory. These points are located through the various technical areas where radioactive materials are handled within the 43 square miles encompassing the Laboratory. Mitigating against the consequences of these releases of trace quantities of radionuclides from the operational facilities are the low

population of the area and the mountainous terrain which provides good air dispersion. Within a 50-mile (80 km) radius of Los Alamos a population of only 170,000 is estimated. The meteorological frequency of atmospheric stability, an estimate of the dispersion capability of the atmosphere, at Los Alamos is approximately 40% unstable (good dispersal), 35% neutral (fair dispersal), and 25% stable (poor dispersal), (Los Alamos 1985).

Emissions of radioactivity into the atmosphere at Los Alamos during calendar year 1986 are listed in Table 1. These emissions reflect the diverse nature of the operations at Los Alamos. The strict control required to hold these emissions to acceptable levels involves various measures that are discussed in the following sections.

(Table 1. Principal Radioactivity Emissions from Los Alamos National Laboratory during 1986.)

ENGINEERED CONTROL SYSTEMS

Order DOE 6430 General Design Criteria (DOE 1985) requires adequate ventilation exhaust clean up systems be installed to control emissions to as-low-as-reasonably-achievable (ALARA) levels. Federal regulations (EPA 1985) require that clean air act standards be observed. Table 2 lists the EPA standards that DOE facilities must observe. These are new standards that were imposed on DOE facilities in 1985 and EPA has the responsibility for their enforcement. The principal sources of emissions and the air cleaning controls provided for them at Los Alamos are described below.

(Table 2. EPA Air Emission Standards for DOE Facilities to Protect Any Member of the Public from Airborne Radiation Exposure.)

Plutonium. While plutonium emissions are perceived as the greatest airborne hazard from operations at Los Alamos they do not present the most difficult control problem. The air cleaning techniques that have been developed and practiced since the inception of plutonium processing during the World War II years have proved to be highly efficient. These techniques involve the proper placement of single, double, or triple stages of High Efficiency Particulate Air (HEPA) filters in the ventilation exhaust

flow paths. In addition the final exhaust blowers customarily discharge the air through a tall stack that may range from 20 ft (6.1 m) to 200 ft (61 m). At DOE facilities where plutonium processes involve large industrial quantities the final exhaust filters may include large beds of sand or fiberglass. Los Alamos quantities involve smaller scale research and development amounts and HEPA filters without final sand or fiberglass filter beds have proved sufficient. Figure 2 depicts a typical HEPA filter ventilation exhaust system for laboratory operations. HEPA filter removal efficiencies are minimal for particulates of a size of the order of 0.1 to 0.3 micrometers Activity Median Aerodynamic Diameter (AMAD). Standards for the use of HEPA filtration require that the filter efficiency be in-place tested and that the particulate removal efficiency be no less than 99.97% for 0.3 micrometer size particles (Burchsted 1976).

(Figure 2. Typical Los Alamos Ventilation Exhaust System.)

The various facilities at Los Alamos where plutonium is handled are all provided with HEPA filter banks except for one operation where filter bags of 95% removal efficiency are used. This operation is the major source of plutonium emissions from Los Alamos but is well below limiting radiation concentration guides (RCG) values at the site boundary. Emissions from its stack at CMR Building Wing 3 measured the highest concentration of plutonium from all laboratory processes during 1986 and are calculated to be approximately 0.03% of the RCG for soluble form plutonium in the unrestricted area (6×10^{-14} Ci/m^3). Because these emissions result in such a small fraction of the most restrictive RCG (soluble form), it is difficult on a cost benefit basis to require that expensive HEPA filters be installed to reduce emissions further. In 1983 it was estimated that it would cost five million dollars to add a bank of HEPA filters to reduce the emissions by another factor of 99.97% (Moore 1984).

Uranium. The HEPA filtration methods that were described for plutonium are also applied to the control of emissions of uranium to the atmosphere at Los Alamos. The operations that handle uranium materials were drastically reduced when the major ^{235}U processing operations at Technical Area (TA) 21

were discontinued in June 1984. Uranium emissions from this source have not reduced to zero primarily because the contaminated ductwork for ventilation exhaust from the area has not been replaced. It is expected that this source of emissions will be eliminated in the future by replacement of the contaminated ducts.

Mixed Fission Products. Analysis of small quantities of irradiated specimens including reactor fuel and diagnostic samples, leads to the possibility of mixed fission product (MFP) emissions from the laboratories involved. HEPA filtration only at the glovebox air exhaust point has provided effective control even in the case where the maximum emissions of MFP occur. The maximum emission source is a ventilation exhaust stack at the radiochemistry laboratory at TA-48.

Mixed Activation Products. The mixed activation products are produced from operation of the Los Alamos Meson Physics Facility (LAMPF) proton linear accelerator. This source of radionuclides has proven to be the one with the greatest potential for exceeding EPA standards at Los Alamos. The problem has been recognized over the past few years and studies of remedial action resulted in modifications to reduce emissions to ALARA levels. A more detailed description of this effort is given later.

Radioiodine (^{131}I). The only detectable source for emissions of this isotope at Los Alamos is the Chemistry Metallurgy Research (CMR) Building hot cell operation where irradiated fuel pin specimens are dissolved and analyzed. The quantity of ^{131}I that becomes airborne is extremely small and no absorption system is required for removal. The major radionuclide in the contaminated air exhausted is plutonium and not ^{131}I therefore HEPA filtration is the clean up system that is provided for the exhaust air from this operation.

Argon. There are two significant sources of ^{41}Ar emissions at Los Alamos. One source is the activation products generated in the beam stop at the LAMPF accelerator and the other is also an activation product from operation of the Los Alamos Omega West research reactor. The LAMPF emissions are discussed later and the ^{41}Ar from the reactor operation is discussed

here. The Omega West reactor is an 8 megawatt homogeneous water-cooled tank-type reactor. It serves as a research tool in providing a source of neutrons for fundamental studies in nuclear physics. The components of the reactor include a 6-foot diameter "thermal column" (so called because its purpose is to reduce the energy of fission neutrons to that of the charcoal blocks that make up the column). Air that is vented from this column to the atmosphere is the only source of emissions that contains detectable activity during routine operation of the reactor. The activity has been identified as ^{41}Ar, a neutron activation product of the normal ^{40}Ar present in air. Any other airborne activity in the effluent is at minimal levels not discernible from natural background. Figure 3 shows the components of the reactor and its ventilation exhaust.

(Figure 3. Omega West Reactor Ventilation Exhaust.)

Phosphorus. The Health Research Laboratory (HRL) at Los Alamos uses milligram quantities of ^{32}P in its biology research programs. The emissions of this isotope are almost negligible but are detected and reported. HEPA filtration of the ventilation air exhausted from the Laboratory hoods is provided because there have been times in the past when trace quantities of ^{239}Pu were also handled in these HRL laboratories.

Tritium. The national defense research programs at Los Alamos require handling significant quantities of tritium. Fusion energy research programs also require handling tritium but in lesser quantities. The major tritium handling facility at Los Alamos releases large amounts of tritium to the atmosphere. During 1986 approximately 6,700 Ci were released from routine operations at this remotely located facility. During operations the ventilation exhaust from this area is exhausted directly to the atmosphere. The bulk of the tritium emitted is in the form of elemental gas which would require expensive catalyst enhanced oxidation for removal. Therefore except for abnormal occurrences the ventilation air from the facility is exhausted directly to the atmosphere. However, if concentration limits are exceeded, the air is exhausted through an Environmental Control System (ECS). The ECS consists of a platinum catalyst bed that

converts $^3H_2$ gas to ^3H_2O. The ^3H_2O is absorbed on a molecular sieve as a control on emissions to the environment. This facility has been in operation over many years and is now slated for replacement. The tritium emission control technology of the replacement facility will be based on experience gained in the Tritium Systems Test Assembly (TSTA) that is a relatively new research facility for handling tritium at Los Alamos (LA 1982). The primary objectives of the TSTA are to demonstrate the fuel cycle for fusion power systems. The TSTA provides a facility where solutions can be developed for the problems presented by large tritium handling systems at future fusion facilities. The point of interest in this discussion is the development of a technology for the safe handling of tritium with no major releases. The basic scheme consists of utilizing state-of--the-art technology for converting hydrogen gas to water which is then removed by solid dessicants. The incorporation of automatic features to deploy clean up systems on demand at preset concentration levels and the radioactivity of tritium are the distinguishing features that extend the technology beyond that of ordinary hydrogen. It is expected that the experience gained in the TSTA will be of benefit in maintaining emissions to ALARA levels in facilities where large quantities of tritium will be handled.

DETECTION AND DOCUMENTATION OF EMISSIONS

The Los Alamos National Laboratory has a program in place to comply with its responsibility to protect the public, worker, and environment from exposure to radioactivity from its operations. The program involves collection, analysis, and documentation of data on all releases of radioactivity.

Throughout its history the Laboratory has maintained surveillance on its effluents and their effects. Currently the Environmental Surveillance Group, HSE-8, is responsible for measuring any radioactivity in the environs of the operations. As part of this effort the Group maintains meteorological stations to determine direction and dispersal of airborne contaminants. It also maintains networks of thermoluminescent dosimeters (TLD) and air sampling stations to verify the effectiveness of engineered barriers and controls at the operating facility. This group issues an

annual report on its surveillance activities (LA 1985). This report includes the site boundary doses calculated from their TLD network, and the stack emissions data described below.

The Laboratory radiation protection groups, HSE-1, HSE-10, and HSE-11 have the responsibility of maintaining state-of-the-art continuous sample collection or monitoring systems for airborne radioactivity in the workplace and in the ventilation exhaust systems. Group HSE-1 also operates a Health Physics Analysis Laboratory (HPAL) where one of the tasks is the weekly analysis of ventilation exhaust stream samples. The HPAL receives all the particulate fiberglass discs and iodine collection charcoal filters for analysis. The 2-1/8 inch filter paper discs used to collect particulates are counted for gross alpha or gross beta. The gross alpha counting is done using a computerized zinc sulfide detector photomultiplier tube system that can handle up to 128 samples simultaneously and which automatically compensates for the background radiation measured daily. The gross beta counting is done in a different computerized system using a gas flow proportional counter, and the gamma counting is done using a high resolution germanium lithium (Ge-Li) drifted detector system whose output is connected to a multi channel analyzer to identify the pulse height energy signatures of the various isotopes. The charcoal filters are analyzed using a NaI scintillation counter except for the LAMPF samples. The LAMPF filter paper discs and charcoal cartridges are subjected to the high resolution Ge-Li gamma spectroscopy for identification of the radionuclides shown in Table 3. QA procedures at the HPAL include requirements for NBS traceability of its sources used for calibration. The details of the computerized counting system at the HPAL are reported in the literature (Vasilik 1978).

(Table 3. Particulate or Vapor Product Isotopes Detected in LAMPF Stack Releases During 1986.)

Group HSE-1 also maintains a data base for emissions and publishes monthly and annual reports of the emissions of radionuclides from all sources. Figure 4 is a diagram that traces the flow of emission data from its sources to its published forms. The principal features of the Los Alamos program for measuring emissions are described below.

(Figure 4. Emission data records management.)

Particulate Radioactivity

As has been previously described particulate airborne radioactivity is generated from those operations where plutonium, uranium, phosphorous or mixed fission products are handled. All particulate sampling for these operations is accomplished by drawing a sample of stack effluent through a fixed collector system containing a 2-1/8 inch diameter filter media disc of fiberglass composition. These discs are collected weekly for analysis at the HPAL. The system includes a weatherized enclosure, a positive displacement air pump connected to a one horsepower electric motor, a float-type rotometer, and a thermostatically controlled muffin fan. Sample flow rates are maintained at 2 or 3 CFM (0.94 or 1.42 L/s) as measured by the rotameter. The flow meter is calibrated quarterly with a standardized venturi gage whose calibration is traceable to the National Bureau of Standards. The error of the sampling flow rate is estimated to be $\pm$ 15%. Figure 5 is a simplified diagram of the sampler units.

(Figure 5. Sampler unit components.)

The sampling methodology practiced is based on DOE guidance (J. P. Corley 1983) and the standards for instrumentation for monitoring radioactivity in effluents (ANSI 1974) and sampling airborne radioactive materials (ANSI 1969). For example the sampling point is selected as close as practical to the release source and, with very few exceptions, a minimum of five duct diameters downstream of any transition that could cause a flow anomoly. In most cases even though HEPA filters are upstream of the sampling point an attempt is made to attain isokinetic sampling. This involves sizing the sample probe cross sectional area so that the fixed 2 or 3 CFM (1 to 1.5 L/s) sampling rate results in equal linear velocities in the duct and in the sampling probe. Isokinetic sampling is important for sampling airborne particulates whose AMAD exceeds 5 micrometers and at Los Alamos that condition would exist only at the very few places where HEPA filters are not provided.

The ventilation exhaust air is typically released to the environment via a tall stack. An important parameter to calculate the total emission from the aliquot sampled is the value of the total volume of air that is exhausted during the sampling period. The stack flow rates are established using a twelve point traverse using a standard pilot tube and an electronic digital micronanometer (EDM). The EDM is calibrated against an inclined gage manometer in order to verify its accuracy. The measurements satisfy the standard EPA method 1 requirements for accuracy (EPA 1984) and are estimated accurate to $\pm$ 20%.

Iodine

Only one exhaust stream at Los Alamos contains detectable amounts of radioiodines. This stack at CMR Building Wing 9 is sampled with activated charcoal cannisters placed downstream of particulate filters. The charcoal cartridges are collected on the same weekly schedule as the particulate filters, and are taken to the HPAL for analysis with a NaI scintillation detector.

Argon

^{41}Ar is monitored continuously on a real time basis at the Omega West Reactor exhaust stack by using a commercial beta-gamma continuous air monitor (CAM). A 2 CFM (0.94 L/s) sample of the stack flow is drawn through a moving belt type particulate filter which is counted in the detection chamber of the CAM. A NaI scintillation detector measures the disintegration of the ^{41}Ar. The ^{41}Ar activity in counts per second is simultaneously displayed on an analog meter on the CAM and also remotely in the reactor control room where the counts are recorded. A Cs-137 test source is used to calibrate the CAM on a quarterly basis.

Tritium

Concentrations of elemental tritium gas in air exhausted to the environment are measured continuously using real-time measurements with tritium monitors either manufactured commercially or built in-house at Los Alamos. Each of these monitors utilizes a flow-through ionization chamber with volumes ranging from 0.1 to 57 liters, depending on the required range, sensitivity, and flow rates. Flow rates through the ionization chambers

range from 0.03 - 3 CFM (0.85 - 85 L/min depending on the chamber size. The ionization chambers are coupled to electrometers for measuring the charge accumulation. The electrometers are either integrating or non-integrating. Either electrometer measures the tritium effluent concentrations, but the integrating electrometers also display the total charge collected during the monitoring interval. Strip chart recorders are used to produce hardcopy records of normal tritium emissions to the atmosphere and to evaluate abnormal emission quantities. Instruments used to measure tritium emissions to the atmosphere are calibrated routinely to NBS traceable standards.

A different analytical method is required for measuring tritium that has combined with water vapor. Passive stack monitors are used for the collection of tritiated water vapor entrained in the ventilation air that is exhausted. These passive monitors employ glycol bubblers in series with a heated precious metal catalyst and additional glycol bubblers. The tritium collected in the bubblers is measured by liquid scintillation counting.

Mixed Activation Products

The principal radionuclides formed from proton and neutron reactions in the LAMPF target cell and beam stop area are gaseous forms of the isotopes of oxygen, carbon, nitrogen, and argon. Real time monitoring of the emissions of these radionuclides to the atmosphere is accomplished using a 5 liter ionization chamber. A sample flow rate through the chamber is maintained at 3 CFM (1.42 L/s) using a float-type rotameter as described previously for the particulates.

An electrometer is used to integrate the accumulated chamber current during the monitoring interval. The concentration of each radionuclide is calculated based on its isotopic fraction, the total integrated charge and the flow rate through the chamber. The isotopic fraction is determined by a spectral analysis. The reduction of the emission of gaseous activation products produced from LAMPF accelerator operation is discussed below.

REDUCTION OF EMISSIONS FROM LAMPF ACCELERATOR OPERATIONS

The LAMPF facility consists of a linear proton accelerator approximately 0.5 mi (800 m) long, designed to produce an 800-million electron volt proton beam with an average beam current of one milliampere. The energetic protons are used to strike a target to produce subnuclear particles called pi mesons, which are of major importance to studies of the fundamental structure of the nucleus. Pi mesons are believed to be the "glue" that holds atomic nuclei together. A consequence of the beam impinging on the target is the generation of activation products from the interaction of the energetic protons with the materials in the target area. Protons transmitted through the target are dissipated on striking a water cooled copper beam stop. Figure 6 shows a cross-sectional view of this area. Activation products are also generated within this beam stop area by interactions with lead shielding blocks, copper and ambient air. The activated air from the beam stop area space is exhausted through a single stage of HEPA filters to the atmosphere via a 60-ft (18 m) stack.

(Figure 6. LAMPF target and beam stop area.

The radionuclides emitted from the LAMPF operation cover a wide range. Proton activation products of the normal isotopes of the ambient air make up nearly 100% of the emissions and consist principally of ^{41}Ar and short lived isotopes of oxygen, carbon, and nitrogen as shown in Table 4. The proton interactions with the copper and lead present in the beam stop area generate a relatively insignificant but detectable amount of particulate and vapor activation products. Most of these products are extremely short lived nuclides with half-lives ranging from several minutes to several days. Gamma spectroscopy measurements of these activation radionuclides have resulted in the identification of 42 radionuclides starting with ^{7}Be and ending with ^{203}Hg. Table 3 shows the particulate or vapor activation radionuclides that were reported for LAMPF accelerator beam operations during 1986.

(Table 4. Gaseous Activation Product.)

From the beginning of beam operations at LAMPF the problem of emissions of the radionuclides formed has been recognized. As the goal of full beam power was more closely reached each year the, emissions increased. The increase of these emissions and the measures taken to reduce them are described below.

Description of LAMPF Emission Control Technology

Air flowing to the LAMPF stack is passed through a single stage of HEPA filtration to remove particulates. There is no technology in place to remove radionuclides in gas or vapor from the air stream. Areas where the air activation products are produced are continuously ventilated, producing a slight negative pressure to overcome connective leakage of the radioactive products into experimental areas. This unfortunately also transports the radionuclides to the stack. Due to the short half-lives of some of the activation products formed, some reduction in the radionuclide release is obtained by decay due to holdup as the air flows from the various sources to the stack. The extent of the reduction will depend on the radionuclides. In the case of oxygen-15 (t 1/2 = 2.0 min), the holdup could reduce the release significantly. In the case of Tritium (t 1/2 = 12.3 yr) and Beryllium-7 (t 1/2 = 53.3d) the holdup would have essentially no effect on the releases. Figure 7 shows a block diagram of the existing LAMPF ventilation exhaust system.

(Figure 7. Block Diagram of the LAMPF Ventilation Exhaust System.)

Discharge Rate History

The release of air activation products from the LAMPF main stack has increased in recent years as programmatic activities and beam intensities have increased. Discharge rates from this stack are shown in Table 5. The average concentrations of the radionuclides in the air are also shown in Table 5. The radionuclides emitted from the LAMPF beam did not present a significant radiation dose until about 1980 when the calculated boundary dose to a hypothetical individual exceeded 10 millirem whole body dose

equivalent for the first time. The calculated dose assumes a hypothetical individual is present continuously 24 hours a day, 365 days a year at the site boundary where the highest dose occurs. As is shown in the table this boundary dose peaked in 1983, remained high in 1984, and dropped considerably in 1985. In 1986 the slight increase over 1985 is difficult to ascribe precisely to correspondingly small changes in the meteorology, background, and isotopes emitted. A parameter that better illustrates the reductions effected by modifications to the beam stop source is the ratio of curies emitted to beam energy (amp-hr). Based on the curies emitted and the Ci/amp-hr ratio the 1984 emissions were at their worst and have been effectively reduced by a factor of approximately 8, both in 1985 and 1986.

(Table 5. Historical Data for LAMPF Emissions.)

Courses of Action

A permanent committee was formed at LAMPF several years ago to review the stack emissions problem. One objective of the committee was to evaluate possible methods for reducing the release of airborne radioactive material. The first remedial measure undertaken was the repair of leaks in the cooling water system. Radiation damage in the isotope production/beamstop region to the cooling lines resulted in leaks which sprayed water through regions of high proton and neutron intensity and also high temperature. Besides placing a heavy burden on the water makeup system, the resulting radioactive water vapor added to the radioactive stack emissions. Repairs to the water system resulted in 172,000 Curies/amp-hr discharge in 1982 compared with 291,000 Curies/amp-hr in 1971. Unfortunately, serious leaks reappeared in the next years. The first control principle for accelerator emissions learned from this experience was to prevent the production of radionuclides through the elimination of leaks from the water cooling system. This requires a continual maintenance effort.

The next reduction possibility considered was to introduce a delay system for the ventilation air exhausted from the beam stop area. The large air flow to the LAMPF main stack, approximately 17,000 cfm (480 m^3/min) makes it very difficult and expensive to utilize any existing

technology to remove the gaseous activation products from the air stream. One realistic approach would be to provide additional holdup time to allow some decay of the short lived radionuclides, as indicated previously. Extremely large air storage volumes would be required to obtain significant reductions in the radionuclide releases. For example, if an atmospheric pressure air storage system having a storage volume of 328,000 ft^3 (9300 m^3) were applied to the air flowing to the LAMPF stack, the additional holdup time provided would be about 19.4 minutes. With this holdup time, the following reductions in specific radionuclide emissions shown in Table 6 would be obtained. This is based on the accelerator programmatic activities of CY 1981. As a result of the increased time available for decay before release, total emissions from the stack would be reduced from about 489,000 Ci/yr to about 102,000 Ci/yr at the 1981 level of programmatic activities.

(Table 6. Reduction of 1981 Specific Nuclides by Increased Decay Time.)

The air storage tank could be of carbon steel construction and located on a concrete pad adjacent to the LAMPF stack. A tank with a storage volume of 328,000 ft^3 (9300 m^3) would be 98.4 ft (30 m) in diameter by about 43.3 ft (13.2 m) high. A block diagram of the system is shown in Figure 8.

(Figure 8. LAMPF Ventilation System with Storage Tank.)

The estimated capital cost for an atmospheric pressure air storage system, with a storage volume of 328,000 ft^3 (9300 m^3), was $4,300,000 (1983 dollars). The estimated operating cost was $90,000/yr. The capital cost of air storage systems of varying size would vary approximately as the eight-tenths power of the size ratio. Annual operating costs would be almost independent of the size ratio. The high cost of this approach precluded further consideration (Moore 1984).

Of various other possibilities considered the one implemented was to isolate the generating area by enclosing it in a steel box approximately 5 ft x 5 ft x 15 ft or 375 ft^3 (12 m^3). This would allow the radionuclides

to decay in place. This concept complemented a plan to rebuild the region in question to make accelerator components more accessible to repair and simplify changing of experimental targets used to study radiation damage. Since the enclosure required a certain number of penetrations that could not be completely sealed, it was necessary to introduce a controllable small amount of ventilation to reduce diffusion of the radioactive gases into experimental areas. This entire multi-benefit project was achieved in early 1985 at a much lower cost of approximately $2,000,000 compared to the $4,300,000 for the large storage volume. The result was an immediate decrease in emissions by a factor of 8 based on the Ci/amp-hr ratio. Obviously, the most expensive solution may not be the best solution! In this instance we were able to effect a straight forward modification that integrated easily with other developmental plans for the facility. In doing so, the source area became less coupled to the ventilation air exhaust and the relatively short-lived radionuclides were trapped for an interval of time significant to permit decay. Another lesson is illustrated here - isolate the activity that is generated.

An additional measure taken was important. Within this box-like structure, wherever possible, air cavities were filled with steel. This is the final and most elementary lesson. Prevent generation of the offending radionuclides.

Another change contributing to reduction of emissions should be noted. In the original construction of the beam stop region, a large amount of lead was utilized as shielding. As design beam currents were approached, the temperatures increased to the point where some lead actually melted. The alchemists dream was finally realized. Significant amounts of radioactive isotopes of gold, platinum, iridium, mercury and antimony appeared in the stack effluent. It was realized that these isotopes were unimpeded by the HEPA filters, but could be collected with charcoal cartridges. For this case, the solution was to remove most of the offending lead, rather than invest in an expensive vapor cleanup system.

The lesson learned in reducing the emissions from operation of the LAMPF accelerator is that correction of design anomolies is the best solu-

tion to the problem. In our case, elimination of water leaks and lead, and provision of containment to trap the air for additional decay of radionuclides proved more effective than installation, operation, and maintenance of costly air cleaning or large scale systems. The desired result was achieved. The boundary dose levels of 48 and 44 mrem/y determined from TLD measurements for 1983 and 1984 were reduced below the EPA Standard of 25 mrem/y by an ample margin. In 1985 and 1986 the boundary doses determined by the same method were 11 and 17 mrem/y respectively as seen in Table 5.

CONCLUSION

The Los Alamos National Laboratory program to detect, measure, control, and document emissions from its operations is reviewed annually for QA considerations and for incorporation of results in the DOE Effluent Information system. The Health and Safety Division at the Laboratory has the responsibility for ensuring that standards are maintained so that the emission quantities are ALARA and reported as accurately as the current state-of-the-art permits. As has been discussed, sampling practices are based on ANSI standards, flow meter calibrations are traceable to NBS standards, duct flow measurements meet EPA criteria, and radiation detector test sources are also NBS traceable.

The introduction of EPA requirements in 1985 for the control of airborne radionuclides did not present any difficulties for Los Alamos operations. The new annual EPA limiting dose of 25 mrem/y at the site boundary from Los Alamos National Laboratory operations has been exceeded only twice. Both annual emissions occurred before the new standards were promulgated and were the result of LAMPF accelerator operations. A continued effort has resulted in emissions at levels well below the EPA standard.

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TABLE 1

Principal Radioactivity Emissions from
Los Alamos National Laboratory during 1986

Radioactivity	Source Operation(s)	1986 Total Curies
Plutonium (^{238}Pu)[a]	Defense Programs	2.07×10^{-4}
Uranium ($^{235}U + ^{238}U$)[b]	Defense Programs	8.44×10^{-4}
Mixed Fission Products	Irradiated Sample Analysis	2.57×10^{-3}
Mixed Activiation Products[c]	LAMPF Accelerator	1.12×10^{5}
Iodine-131	Irradiated Sample Analysis	3.80×10^{-5}
Argon-41	Research Fission Reactor	2.76×10^{2}
Phosphorus-32	Biological Research	6.99×10^{-5}
Tritium	Defense Programs	1.07×10^{4}

[a] Mostly ^{239}Pu with relatively insignificant quantities of other Pu isotopes.

[b] 84% ^{235}U and 16% ^{238}U.

[c] Principally air activation products ^{13}N, ^{16}N, ^{10}C, ^{11}C, ^{14}O, ^{15}O, and ^{41}Ar.

TABLE 2

EPA Air Emission Standards for DOE Facilities
to Protect Any Member of the Public
from Airborne Radiation Exposure

Primary Standards:	
Whole Body Dose Equivalent	25 mrem/y
Critical Organ	75 mrem/y
Alternative Standards[a]:	
Effective dose equivalent for continuous exposure	$\leq$ 100 mrem/y
Effective dose equivalent for non-continuous exposure	$\leq$ 500 mrem/y

[a]These effective dose equivalents are for all sources and all pathways but exclude natural background and medical procedures and are provided as an option for those installations that exceed 25 mrem/y from airborne emissions but can demonstrate through an exhanced surveillance program that they do not exceed the alternative standards.

TABLE 3

Particulate or Vapor Product Isotopes Detected
in LAMPF Stack Releases During 1986

Isotope	Microcuries Dishcarged	Isotope	Microcuries Discharged
BE-7	8.88×10^{3}	CO-58	9.16×10^{0}
NA-22	5.24×10^{1}	CO-60	4.63×10^{0}
NA-24	4.99×10^{3}	BR-77	8.12×10^{2}
SC-44M	6.64×10^{1}	BR-82	2.23×10^{4}
SC-46	4.69×10^{0}	TA-182	4.44×10^{3}
SC-47	8.27×10^{1}	OS-183	6.86×10^{4}
V-48	9.70×10^{1}	OS-185	2.71×10^{3}
CR-51	5.02×10^{1}	IR-188	1.50×10^{2}
MN-52	1.25×10^{2}	HG-195M	9.34×10^{2}
MN-54	1.01×10^{1}	HG-197M	1.71×10^{2}
CO-56	2.16×10^{0}	HG-203	3.30×10^{2}
CO-57	9.85×10^{1}		
		TOTAL Microcuries	1.15E + 05

TABLE 4

Gaseous Activation Product Emissions from
the LAMPF Main Stack for CY 1986

Radio-nuclide	Half Life (s)	Emission (Ci/y)	Concen-tration (Ci/m^3)	RCG[b] Ci/m^3	Concen-tration RCG
^{3}H	3.87×10^{8}	6.07×10^{0}	3.12×10^{-8}	2×10^{-7}	1.6×10^{-1}
^{41}Ar	6.59×10^{3}	3.36×10^{2}	1.72×10^{-6}	4×10^{-8}	4.3×10^{1}
^{10}C	1.95×10^{1}	6.72×10^{2}	3.45×10^{-6}	3×10^{-8}	1.1×10^{2}
^{11}C	1.22×10^{3}	3.92×10^{4}	2.01×10^{-4}	3×10^{-8}	6.7×10^{3}
^{14}O	7.09×10^{1}	5.60×10^{2}	2.87×10^{-6}	3×10^{-8}	9.6×10^{1}
^{15}O	1.23×10^{2}	4.70×10^{4}	2.41×10^{-4}	3×10^{-8}	8.0×10^{3}
^{13}N	5.98×10^{2}	2.08×10^{4}	1.07×10^{-4}	3×10^{-8}	3.6×10^{3}
N_{16}	7.14×10^{0}	3.36×10^{3}	1.72×10^{-4}	3×10^{-8}	5.7×10^{3}

[a] Concentration at stack. There is a dispersion factor of 14,400 to the site boundary.

[b] Radiation Concentration Guides in Table II of Order DOE 5480.1 Chapter XI for the uncontrolled area.

TABLE 5

Historical Data for LAMPF Emissions

	Curies	Ci/amp-hr[a]	Boundary[b]	Dose/kCi[c]
1980	202,000	178,000	12.3	.061
1981	489,000	291,000	17.0	.035
1982	348,000	172,000	12.0	.034
1983	641,000	343,000	48.0	.074
1984	734,000	377,000	44.0	.060
1985	126,900	46,500	10.7	.084
1986	111,600	43,800	16.8	.150

[a]Rates of Ci emitted to amp-hr of beam energy.

[b]Calculated millrem whole body dose equivalent to hypothetical individual assumed at the site boundary.

[c]Ratio of dose to radioactivity emitted at the site boundary.

TABLE 6

Reduction of 1981 Specific Nuclides
by Increased Decay Time

Principal Nuclides	% of Total	Storage[a] Incremental Half Lives	Equivalent Decontamination Factor	Release[b] Without Storage	Release[c] With Storage
^{11}C	36	0.95	1.94	176,000	91,000
^{13}N	7	1.94	3.81	34,200	9,000
^{15}O	56.7	9.56	753.0	277,300	370
^{41}Ar	0.3	0.18	1.13	1,500	1,300
			Total	489,000	101,670

(a)Tank volume of 328,000 ft^3 (9300 m^3) provides additional decay time equivalent to listed half lives for specific nuclide.

(b)Ci released during 1981 LAMPF Operation.

(c)Ci release estimated for 1981 LAMPF Operation if storage tan had been installed.

LIST OF FIGURES AND CAPTIONS